

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re Application of:	)	
William D. Sigworth	:	
	)	Examiner: CAIN, Edward J.
U.S. Application No.: 10/823,953	:	
	)	Group Art Unit: 1796
Filed: April 13, 2004	:	
	)	Confirmation No.: 5455
For: COUPLING AGENTS FOR NATURAL	:	
FIBER-FILLED POLYOLEFINS	)	
	:	
	)	

**MAIL STOP AMENDMENT**

Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

**DECLARATION UNDER 37 C.F.R. § 1.132**

- I, Dr. William D. Sigworth, a citizen of the United States, do hereby declare that:
1. I am an inventor for the above-identified patent application.
2. I have a Ph.D. degree in physical chemistry from Case-Western Reserve University, Cleveland, OH, which was granted to me in 1970.
3. I have worked in fields of physical and polymer chemistry for the Chemtura Corporation and its predecessors for the past 39 years doing product and process development research as well as technical service in the area of additives for plastic and elastomeric polymers.
4. Natural fibers are added to polyolefin resins, e.g., polypropylene resins and/or polyethylene resins, to reinforce the resultant plastics, thus providing a natural fiber-filled polyolefin composition. The addition of the natural fibers increases the tensile modulus of the composition, thereby better simulating natural wood.
5. Coupling agents are added to polyolefin resins to improve dispersion of the natural fiber in the resin and to increase interfacial adhesion between fiber and resin.
6. Coupling agents consisting essentially of a rigid thermoplastic polypropylene homopolymer that are grafted with a polar monomer improve dispersion and increase interfacial adhesion while maintaining the tensile modulus of the natural fiber-filled polypropylene composition.

7. Adding a coupling agent comprising a rubbery copolymer, such as propylene-ethylene copolymer or propylene-ethylene-ethylidene norbornene terpolymer, to a natural fiber-filled polyolefin composition would reduce the tensile modulus or stiffness of the natural fiber-filled composition. This is contrary to the desired effect, i.e., a natural fiber-filled polyolefin composition having a greater modulus or stiffness.
8. To demonstrate the effects of adding coupling agents comprising rubbery copolymers to natural fiber-filled polyolefin compositions, I tested various natural fiber-filled polypropylene compositions and measured the tensile moduli of these compositions. The results of the testing are shown in Table 1.

Table 1  
Effects of Rubbery Coupling Agents on Natural Fiber-Filled Polypropylene

Blend	Natural Fiber, phr*	Polypropylene, phr**	Maleic Anhydride Grafted Polypropylene Homopolymer,*** phr	Maleic Anhydride Grafted Propylene-Ethylene-Ethylidene Norbornene Terpolymer,**** phr	Tensile Modulus, MPa.	Decrease in Modulus, %
1	31.9	66	2	-	2,541	-
2	31.9	63	1	4	1,496	41.2%
3	53.2	44.7	2	-	3,397	-
4	53.2	41.7	1	4	1,946	42.7%

\*Creafill R0083-an alpha cellulose based fiber

\*\* Profax SG-702 PP-a polypropylene homopolymer

\*\*\*Polybond 3200-a homopolymer polypropylene grafted with 1% maleic anhydride.

\*\*\*\*Royaltuf 498-an elastomeric propylene, ethylene, ethylidene nobornene terpolymer grafted with 1% maleic anhydride.

In a second set of experiments, the effects of replacing a portion of a rigid PE coupling agent with a rubbery EPDM coupling agent were studied in LLDPE containing natural filler, particulate starch. Results are summarized in Table 2.

Table 2  
Effects of Rubbery Coupling Agents on Natural Fiber-Filled LLDPE

Blend	Natural Fiber, phr*	LLDPE, phr**	Maleic Anhydride Grafted Polyethylene Homopolymer, *** phr	Maleic Anhydride Grafted Propylene-Ethylene-Ethylidene Norbornene Terpolymer, **** phr	Tensile Modulus, MPa.	Decrease in Modulus, %
1	30	68.9	1	-	993	-
2	30	68.9	0.5	0.5	817	17.7%
3	30	68.9	-	1	642	35.3%

\*Pearl Starch B-a particulate starch from Cargill

\*\* GA501020 LLDPE-a Butene copolymer with MFI of 1.0 from Equistar

\*\*\*Polybond 3029-a homopolymer polyethylene grafted with 1.6% maleic anhydride.

\*\*\*\*Royaltuf 498-an elastomeric propylene, ethylene, ethylidene nobornene terpolymer grafted with 1% maleic anhydride.

9. As shown in Tables 1 and 2, tensile moduli of the natural fiber-filled polyolefin compositions decrease significantly with the addition of coupling agents comprising rubbery copolymers or terpolymers grafted with a polar monomer, e.g., maleic anhydride.
10. The replacement of 20 wt% of maleic anhydride grafted homopolymer with maleic anhydride grafted propylene-ethylene copolymer in a coupling agent would have a significant effect on the essential nature of the resultant coupling agent and would significantly reduce the tensile moduli (stiffnesses) of natural fiber-filled polyolefin, e.g., polypropylene or polyethylene, compositions that employ such coupling agents.
11. I, the undersigned, hereby declare under penalty of perjury that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that statements and the like so made are punishable by fine or imprisonment, or both, under 18 U.S.C. § 1001 and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Application No. 10/823,953  
Atty Dkt. 0176-PA-CIP

Declaration under 37 C.F.R. § 1.132

Dr. William D. Sigworth  
Dr. William D. Sigworth

August 19, 2009  
Date